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**Declarations under Rule 4.17:**

— as to applicant's entitlement to apply for and be granted a patent (Rule 4.17(ii)) for the following designations AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG)

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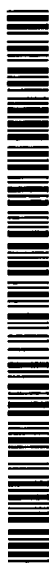
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(54) Title: **UNIVERSAL COMPATIBILIZING AGENT FOR POLYOLEFINES AND POLAR PLASTICS**

(57) Abstract: Product, method and use of a copolymer AB as a compatibilizing agent between a polar thermoplastic polymer C and a polyolefine D. The method of making AB comprise the steps of: a) melting a copolymer (A) comprising a larger amount of methylmethacrylate units and a smaller amount of functional vinyl or (meth)acrylate units wherein functional moiety is selected in the group consisting of (a hydroxyl, a carboxylic, a glycidyl or an amine functional group); together with maleic anhydride grafted polypropylene (B) allowing condensation reaction to occur between the functions under "A" and the anhydride under "B"; b) the condensation copolymer obtained under a) is further melt together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.



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**Universal compatibilizing agents for polyolefines and polar plastics**

Promoting the adhesion between two immiscible polymers has been a concern of  
5 increasing interest. Most useful applications are polymer alloying in very fine  
dispersion of one polymer into the other, and laminated structures and profiles  
obtained by coextrusion or colamination for example.

Various solutions have been proposed to induce some degree of interfacial  
bonding between two polymers exhibiting no or poor mutual adhesion. Among  
10 others, a favorite route is interleaving a third polymeric component whose  
segments provide good spontaneous adhesion with either component  
respectively. Master examples are di-block copolymers made by anionic  
polyaddition [references: S. H. Anastasiadis, I. Gancarz and J. T. Koberstein,  
*Macromolecules* **1989**, *22*, 1449-1453; C. Creton, E. J. Kramer, C.-Y. Hui and  
15 H. R. Brown, *Macromolecules* **1992**, *25*, 3075-3088]. Unfortunately, none of the  
existing commercial block copolymers is suitable to provide enough bonding  
strenght between a polyolefine D such as polypropylene and a polar polymer C  
such as polyvinyl halide, polyvinylidene halide, poly(meth)acrylate, polyester, or  
polycarbonate.

20 It has been found surprisingly that a suitable compatibilizing agent AB of binary  
or ternary mixtures comprising at least one compound of the two above cited  
series of polymers C and D, can be obtained by the following way :

i. A copolymer (A) comprising a larger amount of methylmethacrylate units and  
a smaller amount of functional vinyl or (meth)acrylate units wherein the  
25 functional moiety is characterized as being a hydroxyl, a carboxylic, a glycidyl  
or an amine functional group.

ii. The copolymer A described under "i" is melt processed together with maleic  
anhydride grafted polypropylene (B) allowing condensation reaction to occur  
30 between the functions under "i" and the anhydride under "ii".

iii. The new segmented multiblock copolymer obtained under "ii", is further melt processed together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.

5

Accordingly, the present invention provides a method of making a copolymer AB as a compatibilizing agent between a polar thermoplastic polymer C and a polyolefine D comprising the steps of:

- a) melting a copolymer (A) comprising a larger amount of methylmethacrylate units and a smaller amount of functional vinyl or (meth)acrylate units wherein functional moiety is selected in the group consisting of (a hydroxyl, a carboxylic, a glycidyl or an amine functional group);
- together with maleic anhydride grafted polypropylene (B) allowing condensation reaction to occur between the functions under "A" and the anhydride under "B";
- b) the condensation copolymer obtained under a) is further melt together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.

In a preferred embodiment of the invention, the copolymer A contains 0.02 to 6 (x) mole percent of structural units bearing the functional group, the grafted copolymer B contains from 0.2 to 3 (y) mole percent of maleic anhydride units, the ratio of copolymer A and grafted copolymer B in the multiblock copolymer AB is such that the ratio x over y lies between 10 and 1 and preferably between 6 and 2.

In another preferred embodiment of the invention, the copolymer A contains between 0.1 to 6 mole percent of 2-hydroxyethyl(meth)acrylate and its weight average molecular weight (measured as PS equivalent molecular weight) is between 40000 and 500000 g/mol, preferably between 150000 and 500000 g/mol.

The copolymer A is easily obtained for example by solution polymerization in a suitable solvent such as methylethylketone (MEK), using a thermal initiator.

The present invention also provides a use of copolymer AB as defined above as a compatibilizing agent between a polar thermoplastic polymer C like polyvinyl halide homo or copolymer, polyvinylidene halide homo or copolymer,  
5 poly(meth)acrylate, polyester, or polycarbonate and a polyolefine D as single polymer or in blend with other polyolefines.

Polyolefine D can be polypropylene homo or copolymer with one of more co-monomer and made either with Ziegler-Natta catalyzer or with single site  
10 catalyzer. Polypropylene copolymer can have random or multisequence structure including the reactor made copolymer known as block copolymer.

Polyolefine D can also be a homo polyethylene or a co-polyethylene with zero to 50 wt % of a co-monomer with a number of carbones from 3 to 20.  
15

Endly polyolefine D can be a blend of a polymer D defined above with a polyolefinic rubber like EPDM, butyl rubber, BR, SBR.

Maleic anhydride grafted polypropylene is available in various grades and can be  
20 supplied by DuPont under the trademark FUSABOND®, by Exxon under the trademark EXXELOR®, by Uniroyal under the trademark POLYBOND®, by Atofina under the trademark OREVAC™ for example.

#### Example 1:

25 In a 2 liter stirred reactor, 840 g of methylethylketone, 274 g of methylmethacrylate, 6 g of 2-hydroxyethylmethacrylate (HEMA) and 2.8 g of azobis(isobutyronitrile) are introduced. The oxygen is removed by flushing dry nitrogen in the solution. The temperature is raised to 66 °C. The solution is reacted over 24 hours at 66 °C. An additional 1.4 g of azobis(isobutyronitrile) is  
30 added to the solution and the reaction is carried on during 12 more hours. The

copolymer is recovered by precipitation of the solution in deionized water as non solvent and then dried. The yield obtained is 95 % for the methylmethacrylate and 100 % for the HEMA. The copolymer is characterized  
5 by weight average molecular weight of 152000 g/mol as measured by gel permeation chromatography.

The reactive melt processing leading to the copolymer AB is carried on by mixing 200 g of copolymer A, as prepared following the above description, and  
10 100 g of copolymer B, FUSABOND® MD-353D, on a two roll mill at 180 °C during 30 minutes.

Sandwich elements consisting of a polypropylene sheet (1 mm thick) and a polyvinylidene fluoride sheet (2 mm thick) are bonded with a thin film (100  
15 microns thick) of copolymer AB. The sandwiches are bonded by pressing the above construction during 15 minutes at 150 °C. The adhesion between the two sheets is tested by means of a U-peel test (reference: S. Wu, *Polymer interface and adhesion*, M. Dekker Inc., New York, 1982). A fracture energy (G) of 1607 J/m<sup>2</sup> has been measured.

20

Examples 2 to 5:

In the example 2 to 4, the same experimental procedure as described in the example 1 has been followed. The HEMA amount introduced in the copolymer A is varied (see the following table) as well as the weight average molecular  
25 weight, by means of a transfer agent (dodecanethiol-DDT). The measured fracture energies (G) are given for each example.

30

5

Example number	mol % HEMA in		copolymer A		wt % DDT <sup>a</sup>	Mw
(g/mol) of copolymer A	x/y	G (J/m <sup>2</sup> )				
2	1.68	0.49	94000	2.3	1036	
3	1.76	0.17	222000	2.4	1513	
5	4	5.09	0	172000	5.4	1659
5:without copolymer		-	-	-	-	63

<sup>a</sup>: This percentage is relative to the initial total mass of monomers used in the synthesis of copolymer A.

#### 10 Example 6:

The blending of polyvinylidene fluoride with polypropylene is evaluated with and without using the copolymer AB. In particular, the blend of 19 weight percent of polyvinylidene fluoride, 76 weight percent of polypropylene and 5 weight percent of a copolymer AB (copolymer number 5 in the table presented above) prepared on a two roll mill (roll diameter: 110 mm; temperature: 180 °C; roll speed: 15 rpm; gap: 1 mm) leads to a two phase material presenting particles whose diameter ranges between 3 and 10  $\mu\text{m}$  whereas the blending of polyvinylidene fluoride and polypropylene without copolymer displays particles characterized by a larger diameter, between 15 and 80  $\mu\text{m}$ .

## CLAIMS

1. Method of making a copolymer AB as a compatibilizing agent between a polar thermoplastic polymer C and a polyolefine D comprising the steps of:
- 5 a) melting a copolymer (A) comprising a larger amount of methylmethacrylate units and a smaller amount of functional vinyl or (meth)acrylate units wherein functional moiety is selected in the group consisting of (a hydroxyl, a carboxylic, a glycidyl or an amine functional group);
- together with maleic anhydride grafted polypropylene (B) allowing condensation
- 10 reaction to occur between the functions under "A" and the anhydride under "B";
- b) the condensation copolymer obtained under a) is further melt together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.
- 15
2. Method according to claim 1 wherein the copolymer (A) contains 0.02 to 6 (x) mole percent of structural units bearing the functional group, the grafted copolymer (B) contains from 0.2 to 3 (y) mole percent of maleic anhydride units, the ratio of copolymer A and grafted copolymer B in the multiblock copolymer
- 20 AB is such that the ratio x over y lies between 10 and 1 and preferably between 6 and 2.
3. Method according to claim 2 wherein the copolymer (A) contains between 0.1 to 6 mole percent of 2-hydroxyethyl(meth)acrylate and its weight average
- 25 molecular weight is between 40000 and 500000 g/mol, preferably between 150000 and 500000 g/mol.
4. Use of copolymer AB obtained by the method according to claim 1 to 3 as a compatibilizing agent between a polar thermoplastic polymer C and a
- 30 polyolefine D .

5. Use of copolymer AB according to claim 4 wherein the Polyolefine D is a polypropylene homo or a copolymer with one of more co-monomer.
- 5      6. Use of copolymer AB according to claim 4 wherein the Polyolefine D is a homo polyethylene or a co-polyethylene with zero to 50 wt % a co-monomer with a number of carbone from 3 to 20.
- 10      7. Use of copolymer AB according to anyone of claim 4 to 6 wherein the Polyolefine D is a blend with a polyolefinic rubber.
8. Use of copolymer AB according to Claim 7 with a rubber phase partially or fully crosslinked.
- 15      9. Use of copolymer AB according to Claims 4 to 8 with additive like inorganic filler and well known polymer stabilizers.
10. Copolymer AB obtained by the method according to anyone of claim 1 to 3



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Jean-Marie [BE/BE]; Moulin de Wadeux, 411, B-4654  
Charneux (BE). OUHADI, Trazollah [BE/BE]; Quai  
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Quai Van Beneden, 25, B-4020 Liège (BE).(81) Designated States (national): AE, AG, AL, AM, AT, AU,  
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GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC,  
LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW,  
MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK,  
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IT, LU, MC, NL, PT, SE, TR). OAPI patent (BF, BJ, CF,

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## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Section Ch, Week 199405 Derwent Publications Ltd., London, GB; Class A17, AN 1994-040099 XP002190638 & JP 05 345868 A (KURARAY CO LTD), 27 December 1993 (1993-12-27)	1,10
Y	abstract	1-10
Y	--- "MODIFICATION OF POLYAMIDE BY A CRYSTALLINE POLYOLEFIN AND GRAFT COPOLYMER" RESEARCH DISCLOSURE, KENNETH MASON PUBLICATIONS, HAMPSHIRE, GB, no. 332, 1 December 1991 (1991-12-01), pages 913-917, XP000274621 ISSN: 0374-4353 the whole document ---	1-10
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Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

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\*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>DATABASE WPI  Section Ch, Week 198601  Derwent Publications Ltd., London, GB;  Class A17, AN 1986-004879  XP002190639  &amp; JP 60 233131 A (MITSUI TOATSU CHEM INC),  19 November 1985 (1985-11-19)  abstract</p> <p style="text-align: center;">---</p>	1, 10
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